Sources of Evaluated Data

All subshell parameters are theoretical as opposed to experimental. The basic subshell data and the radiative widths came from the work of J. H. Scofield[6-9]. The nonradiative widths came from the results of M. H. Chen[10-16]. The transition probabilities were evaluated as described in the section,Transition Probabilities and Widths.

Procedures Used to Derive Data

Assumptions and Approximations

This library contains data for subshells that are occupied when an atom is in its neutral, ground state. Data are not included for initially ionized or excited atoms.

The library does not describe the electron that is emitted when the atom is initially ionized. It is assumed that this electron has been explicitly accounted for based on the kinematics of the ionizing event, e.g., photoelectric, electron impact ionization, or internal conversion. It is also assumed that once ionized, the relaxation is independent of how the atom was initially ionized.

The relaxation data in the library is designed to describe the bound-bound radiative and nonradiative transitions following an ionization event. Additionally it is assumed that relaxation only occurs where both radiative and nonradiative transitions are possible.

It is also assumed that the binding energy of all subshells are the same for neutral ground state atoms as for ionized atoms. This assumption has been adopted because at the current time there is no provision to provide complete data describing the binding energy of each subshell of an atom when any of its other subshells is singly or multiply ionized.

Subshell data are provided for Z = 1 through 100; however, transition probabilities are only explicitly included for Z = 6 through 100. Therefore, for Z = 1 through 5 the library only includes a local energy deposit corresponding to the binding energy of an electron in each subshell.

Relaxation data is provided for all subshells that are occupied when an atom is in its neutral ground state. However, transition probabilities are only explicitly included for most subshells. For the outer subshells, the radiative transition probability is very small. Also, the binding energy of an electron is very small so that radiative or nonradiative transitions will lead to the emission of very low energy photons or electrons. In either case these may be considered to deposit their energy locally. Therefore for the outer subshells, the library only includes a local energy deposit corresponding to the binding energy of an electron in each subshell.

Transition Probabilities and Widths

The creation of a vacancy in an atomic subshell initiates a series of complex transitions as the vacancy moves to outer subshells and the atom relaxes back to the stable configuration. There are two types of transitions, radiative and nonradiative. In a radiative transition, a vacancy in one subshell is filled by an electron from an outer subshell with the release of fluorescence radiation, i.e., x-ray emission. In a nonradiative transition, the initial vacancy is filled by an electron from an outer subshell, the available energy given to the removal of an electron from the same subshell or from one further out. This process results in two electron

vacancies. If the initial and second vacancy are in different shells, this is an Auger process. If they are in the same shell, it is a Coster-Kronig transition.

The calculated relaxation data were presented by Scofield[6] and Chen[10] as partial transition rates, S (the inverse of the transition lifetime) and were obtained using Dirac-Hartree-Slater theory. These were converted to partial widths, Γ, through the uncertainty principle, $\Gamma = \hbar S$. Define the partial widths:

Γ_r (i,j) - partial width of a radiative transition, given a vacancy in subshell i moving to subshell j - this results in a single vacancy in subshell j and the emission of an x-ray,

Γ_{nr} (i;j,k) - partial width of a nonradiative transition, given a vacancy in subshell i moving to subshell j and emission of an electron from subshell k – this results in vacancies in subshell j and k and the emission of an electron.

The radiative and nonradiative widths for subshell i are obtained by summing over the terminal subshells

$$\Gamma_r(i) = \sum \Gamma_r(i,j), \tag{1}$$

$$\Gamma_{nr}(i) = \sum \Gamma_{nr}(i;j,k), \tag{2}$$

and the total width is

$$\Gamma_t(i) = \Gamma_r(i) + \Gamma_{nr}(i). \tag{3}$$

The two transition probabilities of interest are the radiative

$$f_r(i,j) = \Gamma_r(i,j)/ \Gamma_t(i), \tag{4}$$

and the nonradiative

$$f_{nr}(i;j,k) = \Gamma_{nr}(i;j,k)/ \Gamma_t(i). \tag{5}$$

The energy into the x-ray is

$$E(\gamma) = E(i) - E(j) , \tag{6}$$

and into the emitted electron is

$$E(e) = E(i) - [E(j) + E(k)] , \tag{7}$$

where E(i), etc., are the subshell binding energies.

Dirac-Hartree-Slater calculations,however, seriously overpredict the strength of Coster-Kronig transitions that required modification to the data. Hubbell[17] gives formulas to predict the fluorescence yield for the K, L, and M shells for any element. The basic Z dependence is a power series, in particular, here one encounters the familar Z^4 dependence for radiative yield. The formulas of Hubbell were extended to predict the fluorescence yields for individual subshells of the K, L, M, N, and O shells. In each case, the yield is expressed as a power series in Z^4, normalized to the yield predicted by the data of Scofield and Chen for high Z elements and adjusted for the opening of individual subshells of each shell. The fact was used that generally the uncertainity in the nonradiative widths is larger than that for the radiative

widths. Furthermore, the uncertainty in the Coster-Kronig widths is larger than that in the Auger widths. Thus to adjust the fluorescence yield i.e., what is described below as the enhanced yield, to agree with atomic number (Z) systematics, the nonradiative transitions probabilities, in particular the Coster-Kronig transition probabilities, were modified.

Number of Transitions

Radiative transitions numbered some 7667 data points and nonradiative transitions totaled 90,513 data points. In order to make the data in EADL consistent, transitions were only considered where there was data to describe both radiative and nonradiative transitions.

Transition probabilities were only included for Z = 6 through 100 and only from the subshells of the K, L, M, N shells and some O subshells. In all other cases (Z = 1 through 5 and subshells of the O, P, and Q shells) the emitted photons and electrons will have very low energies and it is assumed that they deposit their energy locally, i.e., in all these cases EADL only includes a local energy deposit equal to the binding energy.

Energy Deposition

Atomic relaxation back to stability is important as it is a source of electrons and x-rays. The available energy for the relaxation is equal to the binding energy of the electron originally ejected, which can be over a hundred keV. The manner in that this energy is partitioned between photons and electrons can be crucial to energy deposition.

For a heavy element, the number of transitions involved in the cascade back to neutrality can be very large. In the case of uranium with an initial vacancy in the K shell, over 1000 nonradiative and close to 100 radiative transitions are possible. A very large population of such atoms would exhibit a spectrum with many lines. The average number of transitions a single such atom would actually experience, is much less: just over 10 nonradiative and about 8 radiative transitions.

A computer code[18] was written to calculate such average quantities. Assume a single vacancy exists in a given subshell. This vacancy then is partitioned and propagated outward through the transitions probabilities, creating essentially a cascade of vacancies. As there are no transition probabilities to the continuum, the vacancies finally stop in the outer subshells where they contribute to "local" deposition. The number and energies into the photon and electron fields is also determined. By energy conservation for a specific subshell, the binding energy equals the average total energies into the photons and electrons plus the local deposition.

A basic assumption made here is that changes in the binding energies from those of the original neutral atom are neglected. This approximation can result in negative (a few eV) binding energy differences and corresponding erroneous negative electron energies. This actually affects very few of the transitions. To correct for this, if the ejected electron's energy was calculated as less than zero, it was set equal to zero. The contribution of this difference from zero (of negative magnitude) was then added to the local deposition, which still insures overall energy conservation.

Fluorescence Yields

The same computer program[18] that calculated energy deposition calculates fluorescence yields. There are three different yields that can be defined for a subshell:

1) Direct - For a single initial vacancy in subshell i, this includes all photons emitted due to transitions filling this vacancy. In terms of the widths defined by Eqns. (1) - (3), the direct yield, is defined as $\omega_i = \Gamma_r(i)/\Gamma_t(i)$, which corresponds to Krause's[19] definition of ω_i .

2) Enhanced - For a single initial vacancy in subshell i, this includes all photons emitted due to transitions filling either the initial vacancy or filling any other vacancies within the *same shell* that have been created due to the initial vacancy. The enhanced yield corresponds to Krauses's[19] definition of the effective yield, υ_i .

These other vacancies can be created by radiative or nonradiative (Coster-Kronig) transitions between subshells of the same shell. Compared to the direct yield, these additional photons emitted from the same shell will enhance the observed photon yield. Since the binding energy of different subshells in the same shell are very similar, it is experimentally difficult to distinguish between the direct and the enhanced photon yields.

3) Total - For a single vacancy in subshell i, this includes all photons emitted due to transitions filling either the initial subshell vacancy or filling any other vacancies within *any subshell* that have been created due to the initial vacancy.

Compared to the enhanced yield, the total yield may include additional photons but generally, these will be of lower energy that is more readily distinguished from the direct and enhanced yields in experimental observations.

From the above definitions, one has: total ≥ enhanced ≥ direct .

Accuracy of Data

By comparing subshell parameters from a number of different sources, it can be seen that there is still a disagreement of about 1% between the binding energies. For use in applications, particularly coupled electron-photon transport, knowing the exact binding energies is not as important as ensuring that the same binding energies are used throughout. Therefore, Scofield's subshell parameters[6] are consistently used for both atomic, photon, and electron data.

Based on the calculations of Chen, [10-16] the Auger (nonradiative) widths for an inner shell vacancy are known to better than 15% if the inner shells do not decay by Coster-Kronig or super Coster-Kronig transitions; for these transitions, the widths can be too large by a factor of 2. These uncertainties directly affect the competition between radiative and nonradiative yields (e.g., the fluorescence yield).

The K and L shell radiative rates from Scofield's [6-9] calculations are accurate to about 10%. For outer subshells with transitions under 100 eV, inaccuracies of 30% would not be surprising.

Associated Libraries and Availability of the EADL

EADL only contains the data necessary to describe the relaxation of ionized atoms. In order to perform coupled photon-electron calculations, in particular to describe events that lead to ionization, two additional libraries are available.

1) The Livermore Evaluated Photon Data Library (EPDL)[2], to describe the interaction of photons with matter.

2) The Livermore Evaluated Electron Data Library (EEDL)[4], to describe the interaction of electrons with matter.

All three of these libraries are available in the ENDL format.[1] It should also be noted that the results presented here describe EADL as of July, 1991. Because of recent reevaluation, the present results regarding x-ray (fluorescence) emission, electron emission, and energy deposition supercede the results reported in our earlier publications.[2,20] The subshells that are present in both the EADL as well as in the other two libraries are listed in Table 1.